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## MOIST EXPANSION OF CERAMIC MATERIALS

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It is demonstrated that introduction of wollastonite in the composition of ceramic mixtures promotes an increase in the RO content, a decrease in the ratio of  $\text{Al}_2\text{O}_3 + \text{SiO}_2 : \text{RO}$ , and neutralizes the negative effect of  $\text{R}_2\text{O}$  on the moist expansion of ceramic tiles. Likewise pyrophyllite can contribute to a decrease in the expansion of tiles but is less efficient than wollastonite.

The moist expansion of facing tiles is a quality parameter determining their service properties. Ceramic tiles in a moist environment expand in size, which produces additional stresses in glazes. When such stress exceeds the elasticity of the glaze, crazing arises in the glaze.

According to the standard perceptions, moist expansion of fired material depends on its phase and chemical composition. Moist expansion of ceramics consists in the expansion of the crystalline, vitreous, and amorphous phases. The highest degree of expansion is observed in the amorphous phase, and the lowest in crystalline materials. The effect of the vitreous phase on moist expansion is determined by its chemical composition and can vary significantly.

In the general case, moist expansion decreases, as the content of the crystal phases and alkaline-earth oxides in the fired material increases and the content of the amorphous phase and alkaline oxides ( $\text{R}_2\text{O}$ ) decreases.

Moist expansion is largely determined by the reactions with specific ceramic phases, mostly with the vitreous phase [1]. Introduction of alkaline-earth oxides or alumina in the

composition improves the hydrolytic stability of the glass. A decrease in moist expansion is also achieved by introduction of quartz sand or other silica materials in the mixture [2]. However, the main reasons determining the degree of moist expansion have not yet been established, and no simple methods for calculation of the compositions of mixtures ensuring a preassigned level of moist expansion of ceramics have been proposed, which induced the authors to carry out the present research.

The moist expansion of ceramic materials can be estimated by two methods: the statistical and the dynamic method [3].

The use of the statistical method makes it possible to precisely measure the length of the sample before and after its treatment with water vapor in an autoclave. The data on the natural expansion of ceramic materials in the environment are of special interest. These data can be obtained with a dilatometer. A ceramic sample with a certain degree of swelling selected for testing is placed in a dilatometer and heated twice. On the first heating, a curve with a small slope in the low temperature region is obtained, and on the second heating, the dilatometric curve has a normal course. The vertical

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TABLE 1

Component	Weight content, %, in the mixtures													
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Argillaceous component of gravitation tails of zircon-ilmenite ores	50	50	50	50	50	50	50	50	50	—	—	—	—	—
Zhana-Daurskoye clay	—	—	—	—	—	—	—	—	—	60	60	60	55	55
Light fraction ashes	50	35	30	25	—	—	30	20	10	—	40	—	—	—
Belgorod feldspar concentrate	—	—	—	—	50	30	—	—	—	40	—	25	25	20
Wollastonite	—	15	20	25	—	20	—	—	—	—	—	15	20	25
Pyrophyllite	—	—	—	—	—	—	20	30	40	—	—	—	—	—

distance between the two curves corresponds to the degree of moist expansion of the sample [3].

Based on this principle, the natural expansion of different ceramic compositions was measured (Table 1). The calculated chemical composition of the ceramic mixtures is given in Table 2.

The authors assumed that moist expansion is fully reversible [3]. The problem of "aging" of ceramics as a result of moistening is insufficiently described in literature, therefore we assumed, the same as the authors in [4], that moistened ceramic material can contain two types of water: weakly bound and strongly bound. The content of weakly

bound water is below 0.01%, and the amount of strongly bound water depends on the type of moistening and varies from 0.2 to 1.1%. Heating of the sample in the dilatometer at a rate of 4°C/min ensures complete dehydration of the material at a temperature of 100°C.

It was found that the volumetric variations in samples of the analyzed compositions virtually cease at the temperature of 400°C. The least significant changes in volume were observed in the samples of compositions 12–14.

It is known [5] that moist expansion of the mixtures containing alkaline additives exceeds the expansion of the mixtures with alkaline-earth additives.

The performed experiments indicated that the least perceptible moist expansion was exhibited by ceramic materials containing wollastonite based on Zhana-Daurskoe kaolin clay (compositions 12–14). Replacement of wollastonite with pyrophyllite (compositions 3, 4, 7, and 8) and replacement of feldspar with ashes causes an increase in the level of moist expansion. With an increase in the CaO content, the expansion decreases, and with an increase in the R<sub>2</sub>O content, it increases.

The analysis of samples of tile mixtures produced by some foreign companies (Italy, Bulgaria, Yugoslavia, etc.) revealed their predominantly alkaline-earth composition [4]. The moist expansion of these samples does not exceed 0.03–0.05%.

Since an amorphous phase is usually formed on thermal decomposition of argillaceous minerals, we made an attempt, the same as the authors in [2], to take into account the dependence of moist expansion on the ratio between the argillaceous minerals and RO and RO<sub>2</sub> which form part of the composition of ceramic mixtures used in ceramic tile production. As distinct from the authors of [2] who studied ceramics based on natural materials, we investigated ceramics contain-

TABLE 2

Composition	Weight content, %					
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	R <sub>2</sub> O
1	60.70	19.40	6.03	2.31	1.93	4.84
2	58.48	16.85	5.26	8.02	1.71	3.74
3	57.74	16.27	5.04	10.01	1.63	3.31
4	57.01	15.47	4.78	12.07	1.57	2.93
5	66.48	17.94	2.02	1.38	0.77	5.90
6	61.21	15.40	3.43	9.38	0.98	3.90
7	58.73	22.89	4.94	1.41	1.41	3.35
8	57.75	24.88	4.44	1.52	1.17	2.00
9	56.68	32.51	3.90	1.24	0.92	1.66
10	71.17	16.22	1.99	2.35	0.98	4.19
11	65.45	17.41	4.81	2.73	2.00	3.33
12	67.21	14.32	2.08	7.60	0.95	2.73
13	66.16	14.55	1.97	9.61	1.10	2.74
14	64.87	13.71	2.00	11.69	1.14	2.25

TABLE 3

Composition	Weight content, %			R <sub>2</sub> O	Component ratio, %		Moist expansion in tiles, %
	SAM	Al <sub>2</sub> O <sub>3</sub> + SiO <sub>2</sub>	RO		SAM : RO	Al <sub>2</sub> O <sub>3</sub> + SiO <sub>2</sub> : RO	
1	25.0	70.66	4.24	4.84	5.90	16.67	0.32
2	25.0	75.93	9.73	3.74	2.57	7.80	0.14
3	25.0	74.34	11.64	3.31	2.15	6.39	0.09
4	25.0	72.80	13.64	2.93	1.83	5.34	0.06
5	25.0	84.42	2.25	5.90	11.62	39.27	0.35
6	25.0	76.62	10.36	3.90	2.41	7.40	0.10
7	25.0	81.98	2.82	3.35	8.86	29.07	0.16
8	25.0	85.93	2.69	2.00	9.29	31.93	0.13
9	25.0	83.28	2.16	1.66	9.40	38.56	0.12
10	30.0	87.39	3.33	4.19	11.57	26.24	0.28
11	30.0	84.41	4.73	3.33	6.34	17.85	0.26
12	30.0	81.53	8.55	2.73	3.51	9.54	0.06
13	27.5	79.70	10.71	2.74	2.57	7.44	0.04
14	27.5	77.76	12.83	2.25	2.14	6.06	0.02

ing waste from non-ferrous metallurgy and the power industry. Table 3 shows the values of moist expansion in tiles fired at the temperature of 1000°C depending on the content of argillaceous minerals, RO, R<sub>2</sub>O, and the ratio between the sum of argillaceous minerals (SAM) and the RO content (RO = CaO + MgO) in the fired material.

It can be seen that moist expansion of facing tiles mostly depends on the content of RO, R<sub>2</sub>O, the ratios of SAM : RO and Al<sub>2</sub>O<sub>3</sub> + SiO<sub>2</sub> : RO. The tiles with the highest RO content and the highest ratio of Al<sub>2</sub>O<sub>3</sub> + SiO<sub>2</sub> : RO exhibit the highest degree of expansion (composition 5), the only exceptions are the tiles containing pyrophyllite (compositions 7 – 9). Compositions 7 – 9 have a high ratio of Al<sub>2</sub>O<sub>3</sub> + SiO<sub>2</sub> : RO, and yet the moist expansion of these tiles is lower than that of the tiles made of compositions 1, 10, and 11. This is probably related to the higher content of Al<sub>2</sub>O<sub>3</sub> in pyrophyllite (34.88%). The lowest expansion is exhibited by tiles with a relatively low content of R<sub>2</sub>O (compositions 4, 12 – 14). At the same time, these tiles have the highest content of RO (except for compositions 2, 3, 6, and 7).

In investigating accelerated firing conditions, the negative role of R<sub>2</sub>O is manifested in compositions 1, 5, 10, and 11, i.e., the mixtures in which pyrophyllite and wollastonite are absent. Wollastonite eliminates the negative effect of R<sub>2</sub>O on moist expansion to a greater extent than pyrophyllite does. Apparently, alkaline oxides do not react to wollastonite and pyrophyllite at the temperature of 1000°C and facilitate dissolution of amorphous products of decomposition of the argillaceous minerals in the melt. In this case, wollastonite and pyrophyllite develop a dense skeleton impeding alteration of the previous volume. The resulting liquid phase in-

tensifies the condensation of new crystal phases, and part of the R<sub>2</sub>O gets bound in feldspars. Subsequent crystallization of the mixture on cooling of the facing tiles securely binds the needles of the crystals to each other. Apparently, all this neutralizes the negative effect of R<sub>2</sub>O on moist expansion of tiles.

In this way, introduction of wollastonite in the composition of ceramic mixtures produces an increase in the RO content, a decrease in the ratio of Al<sub>2</sub>O<sub>3</sub> + SiO<sub>2</sub> : RO and, consequently, neutralizes the negative effect of R<sub>2</sub>O on moist expansion. In the same way, pyrophyllite facilitates a decrease in the expansion of ceramic tiles, but not as effectively as wollastonite. The effect of wollastonite on the decrease in moist expansion is most explicit in facing tiles made of the Zhana-Daurskoe kaolin clay.

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